# DETONATION STRUCTURE IN CONDENSED PHASE EXPLOSIVES

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INTRODUCTION: A detonation may be regarded as a strong shock wave coupled with an immediately-following exothermic reaction. Classically, detonations have been considered to be one dimensional and steady-state relative to the leading shock wave. The one-dimensional steady-state models are quite useful in predicting detonation pressures and shock and particle velocities. However, it is now known that the mechanism whereby a self-sustaining gas-phase detonation propagates with steady gross features involves an intrinsic, multi-dimensional nonsteadiness on a scale commensurate with the reaction zone thickness (1). The major structural features for a number of gas phase systems are reasonably well understood (2). The nonsteady behavior is manifested primarily through the occurrence of weak shock waves with associated Mach stem configurations which propagate across the detonating front as the front processes unreacted material. The main features are portrayed schematically in Figure 1. The solid lines represent shocks, the dashed lines represent Mach stem trajectories, and the dotted lines represent slipstreams. The directions of propagation of the shock waves are indicated by arrows. The velocity of the lead shock is higher than the Chapman-Jouguet (CJ) value immediately after a collision of transverse waves, and decays to a value below the CJ state prior to the next collision. The CJ value corresponds to the average frontal velocity, and is what one would predict using a one-dimensional steady-state theory. The pressure ratio across the transverse shocks is $\sim$ 1.3 for most gaseous detonations. This ratio is constant within experimental accuracy throughout the detonation process and is apparently independent of chemical system and initial pressure (3). The transverse wave strength increases during detonation failure and has a value greater than 1.3 under limit conditions near failure. Ample evidence exists to support the one-dimensional (CJ and Zeldovitch-von Neuman-Doring) models as adequate descriptions of the gross wave properties of nearly planar, steady detonations. However, the transverse wave strength appears undiminished as all spacing decreases, which indicates that the detonation is not approaching a one-dimensional structure (4). Furthermore, the

one-dimensional models are not well-suited to address questions of failure and other nonsteady effects.

The purpose of this investigation was to extend some of the results and knowledge of nonsteady effects in the gas phase to liquid and solid explosives, where the physical and chemical conditions are extremely different. In commonly studied gas phase detonations, the pressures are of the order of one bar ∿ one atmosphere, and the shock tube may be treated as a rigid wall. Furthermore, the gaseous systems are usually highly diluted with inert gases, which lead to very large reaction zones. In liquid and solid phase explosives, the pressures generated are measured in hundreds of kilobars (where each kilobar is one thousand atmosphere). Diluents used to enlarge the reaction zones in liquids in general participate in the reactions, and, for solids, the intrinsic physical heterogeneity of the material generates many shock interactions. Thus, it is of considerable interest to determine how general to detonation phenomena transverse structure is and how the structure is affected by the conditions peculiar to liquid and solid explosives.

EXPERIMENTAL TECHNIQUE: Experiments were designed to duplicate as closely as possible conditions extant in studies of gas phase detonations in rectangular shock tubes. The experimental apparatus for liquid studies is depicted in Figure 2. Container walls were made of brass, machine screwed together and sealed on the outside with a commercial sealant. The large sides were backed by armor plates, providing a total metal thickness on each side of approximately 0.5m. This protected the firing chamber and aided preservation of the recording plates. The inner surface of all recording plates were machined and polished to a mirror finish. The plates on which the transverse wave trajectories of liquid phase detonations were recorded were electroplated with 20.013mm Ag coatings. Best results were obtained with unbuffed, matte finish coatings. The containers were filled with the liquid explosive, which was detonated by means of Dupont "Detasheet"*. Liquids studied were nitromethane-acetone and nitromethane-ethanol solutions.

Configurations for solid explosive experiments were identical to those for the liquids, except that electroplating was found to be unnecessary. Charges (TNT and TNT-Al mixtures) were cast in place. In some shots, the castings were moved in order to break adhesion between the casting and the metal. No difference in results was observed. The above configurations were strictly adhered to except for one series of experiments using partial confinement. Here, experiments using cast solids were performed with just the recording plate, and with recording plate and two contiguous sides present.

*E. I. Dupont de Nemours Co., Wilmington, Del.

RESULTS AND DISCUSSION: A typical record of transverse wave trajectories generated by passage of a time-average steady detonation in a nitromethane: acetone 80:20 (volume) solution is shown in Figure 3. The arrow indicates the direction of propagation of the detonation. The average cell width, measured as the distance between two trajectory intersections in a plane perpendicular to the detonation frontal motion, is 0.6mm. This result is slightly smaller than that determined by Dremin, who obtained a 1.2mm spacing by observing self-luminosity traces on a smear camera record (5). It is found that the cell size increases with distance of the cells from the plane of initiation. At the plane of initiation, the cell size is too small to observe. This result is entirely analogous to gas phase results where the cell size is small upon initiation and grows to a steady state size. Time-average steady conditions are reached a few cm away from the plane of initiation. Near the container walls, wave trajectories diverge slightly, indicating rarefaction influences. Angles and spacing measurements were therefore obtained from the center of the plates, where divergent flow effects are minimized. The average half-angle for the nitromethane-acetone system is found to be $37.5^{\circ}$. If one assumes that the leading shock velocity is equal to the CJ velocity and that the transverse waves are oriented approximately parallel to the charge axis (which assumptions have been shown to be very good approximations in the gas phase) and that the explosion products obey a polytropic aquation of state with $\gamma=3$, the pressure difference across the transverse waves is $\sim$ 81 Kbar. The abcve calculation is clearly inaccurate, but should yield reasonable order-of-magnitude results. Thus, the pressure difference across the transverse waves is quite large.

Gas phase studies have shown the transverse wave strength and intersection angles to be independent of diluent concentration and chemical composition. The cell spacing, however, is a function of both of these. Dremin has proposed that the cell size, for liquid explosives, is a function of activation energy ($\varepsilon$) and heat of explosion (Q)(7). Nitromethane is cited as an example of an explosive with a rough front, as it has a relatively large $\varepsilon$ but a small Q. Tetranitromethane has a smaller activation energy and about the same Q, and is cited as an example of an explosive possessing a smooth front. Dremin's hypothesis is at variance with results of gas phase studies, which yield an increased spacing with reduction of Q. Thus, if true, this hypothesis represents an important difference between behavior in gaseous and liquid systems. In order to test its verity, experiments were run with nitromethane solutions possessing considerably different heats of explosion. The two diluents chosen were acetone and ethyl alcohol, both of which are completely miscible with nitromethane. Acetone has an energy of formation of -615 Kcal/mole and ethanol has a value of 966 Kcal/mole (8). The heat of formation of nitromethane, for comparison, is -212 Kcal/mole (8). For solutions with 20% (vol.) diluent, the resulting heats of formation per unit mass are -512 cal/gm for nitromethane-acetone and 152 cal/gm for nitromethane-ethanol,

neglecting heats of solution. To check that these values represent significant energy changes in terms of detonation parameters, an equilibrium thermodynamic one-dimensional calculation of the CJ pressures was made using the TIGER code (9). The calculated CJ pressures for nitromethane, nitromethane-acetone, and nitromethane-ethanol are 148 Kbar, 131 Kbar, and 181 Kbar, respectively. Experiments on the two diluted solutions were run with identical experimentation, and spacing and trajectory angles were obtained from a region where the detonation was time-average steady. Spacings and angles were identical within the trends of precision for equal diluent concentration. With increased dilution, spacing also increased. The results for these systems are consistent with gas phase observations, but are inconsistent with Dremin's $\epsilon$-Q hypothesis. Apparently, the explosion energy magnitude has little effect upon the spacing and trajectory angles and, hence, transverse wave strength.

The importance of transverse waves to the propagation of a detonation is illustrated by the wave interaction pattern of a failing detonation in a nitromethane: acetone 70:30 (vol.) solution (Figure 6). As the detonation failed, the cell size grew rapidly Collision of transverse waves caused re-initiation of detonation until the overall pressure was too low. In the photograph, one cannot readily discern the Mach stem intersection points, although microscopic examination usually makes them apparent. A few intersection points are indicated by the dots in Figure 6. Note that the resulting pressure and temperature after collision was still quite low, as an induction distance is noticable before detonation broke out. The detonation was then apparently overdriven, as evidenced by the formation of very fine cells which grew as the detonation decayed. Thus, the concept of a multiple shock collision, re-initiation, overdriving, decay sequence in a detonation is strongly supported by these patterns. The interaction patterns are clearly quite complex, and little quantitative information could be gleaned from these experiments.

It is reasonable to expect that transverse wave mechanisms would be important in liquids, since the liquid phase is quite homogeneous. However, solids possess a high degree of heterogeneity, with grain structure size of the order of magnitude of the reaction zone thickness. To investigate the existence of transverse waves in detonating polycrystalline solids, experiments were performed upon cast TNT, using setups identical to those used in liquids studies. The average particle size of the cast TNT was $\sim$ 0.2mm. A photograph of a typical pattern for cast TNT confined in all four sides is shown in Figure 4. Ablation of the surface by the expanding products partially obscures the pattern, but transverse wave trajectories are clearly evident. The direction of propagation of the detonation is indicated by the arrow. The average cell width for cast TNT was found to be 1.5mm in the time-average steady region. The average trajectory angle with respect to the

normal to the detonation front was 34°. A calculation performed in the same manner as for the liquids yielded a transverse wave pressure difference of 145 Kbar. Thus, for TNT, the transverse waves represent sizeable pressure perturbations.

Strehlow and Barthel have advanced an acoustic amplification mechanism to explain the origin and spacing of transverse waves in gaseous systems (10). In this theory, it is assumed that the detonation couples strongly with the acoustic modes of the tube. If this theory is correct, one should also observe transverse waves under weak or no confinement, with the detonation coupling with the acoustic modes of the bar of explosive. Experiments were performed using cast TNT with dimensions and geometry identical to those in previous experiments except that one or three walls were removed. For both types of experiments, an increased spacing was observed. In the case of the casting confined only by the recording surface, a herringbone pattern was observed (Figure 5). Close examination revealed that trajectories which appeared to be generated at the sides of the charge were very weak and difficult to observe, but grew in strength as they progressed to the center. These results are consistent with the acoustic amplification mechanism. Note that a mechanism involving the reflection of transverse shocks at the boundaries of the charge is apparently unnecessary.

Experiments in combustion instability of solid phase rocket motors have shown that introduction of aluminum into the propellant was effective in damping high frequency instabilities (11). The damping was believed to be due to the formation of solid $Al_2O_3$ particles. Aluminum particles approximately 0.015mm in diameter were introduced into TNT to determine the effect upon transverse waves. It was found that a 20% (weight) Al content reduced the amplitude of the transverse waves to the extent that they left almost indiscernable trajectories. No effect was observed on either cell spacing or trajectory angles. The damping effect of the Al on the transverse waves is consistent with either an acoustic mechanism or a shock interaction mechanism. One important effect of Al addition is to greatly increase the time over which heat is released once the material is processed by a shock. Thus, the fact that the frequency of the waves and trajectory angles were unchanged is an indication that the (endothermic) induction zone processes are more important in determining the above parameters than is the exothermic portion of the reaction zone.

CONCLUSIONS AND SUMMARY: It is clear from these studies that the transverse wave interaction phenomena observed in gaseous detonations are also of considerable importance in detonation of liquid and solid phase explosives. The experimental technique utilized above is amenable to the generation of quantitative data regarding transverse wave trajectories and cell sizes. The cell sizes and average trajectory angles of nitromethane solutions are

apparently independent of the total explosion energy. This result is consistent with gas phase observations, but inconsistent with Dremin's hypothesis. The addition of large amounts of aluminum to cast TNT causes an apparent reduction in transverse wave amplitude, but has no noticable effect upon spacing or trajectory angles.

Several directions for further research are indicated. In order to properly study transverse waves in liquids, a liquid explosive which generates highly regular and symmetric cells is required. Once such a system is obtained, a systematic study of the effects of confinement, geometry, etc., upon the detonation structure and nonsteady behavior can be made. Studies of transverse wave phenomena in solid explosives clearly need to be extended to other systems. Price has suggested that solid explosives can be divided into two groups: those that behave more ideally at low porosities and those which behave more ideally at high porosities (12). It is also suggested that the differences arise from the dominance of homogeneous reaction in the former and heterogeneous processes in the latter. It would be valuable to study the transverse wave behavior for members of the two groups, with the goal of obtaining a link between detonation behavior and transverse structure, and the interaction of explosive materials with the confinement systems. Such information is required for the understanding and control of munitions behavior, where the explosive-confinement interaction, its relationship to failure behavior, and steady state detonation parameters are, at present, only slightly understood as a result of extensive, and expensive, empirical testing for each specific system.

This work forms an integral part of the comprehensive program on explosives research being conducted at the Ballistic Research Laboratories and the Picatinny Arsenal under the AMC Explosives Research Program.

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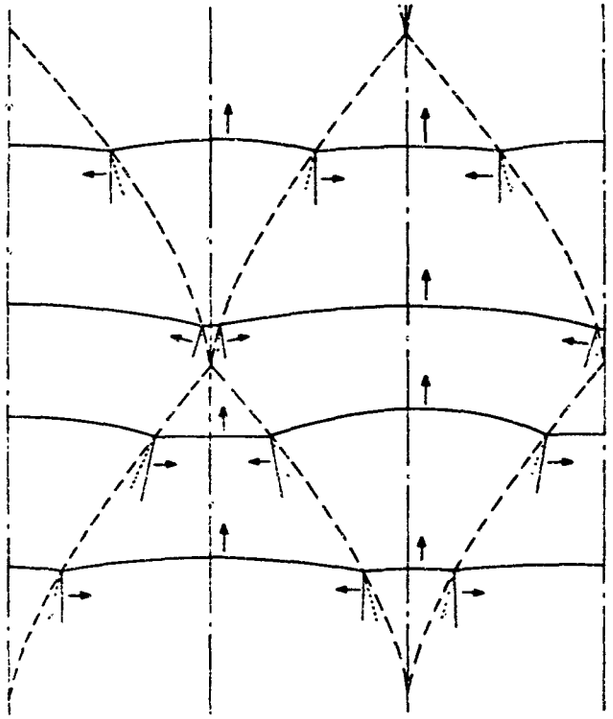

Figure 1 Schematic Representation of Gas Phase Detonation

Figure 1. Schematic depiction of gas phase detonation. Solid lines represent shocks, dashed lines represent Mach stem trajectories, dotted lines represent slipstreams.

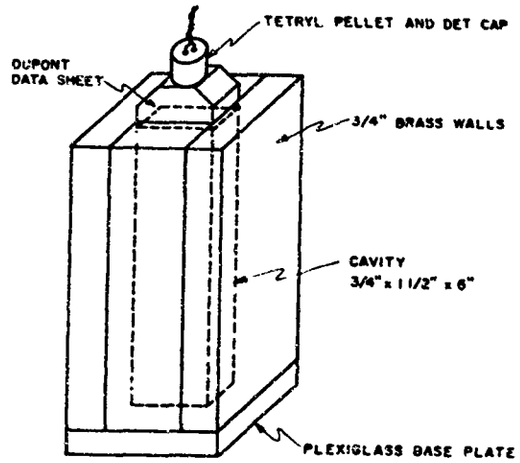


Figure 2 Experimental Setup for Liquids

Figure 2. Experimental apparatus used for transverse wave studies.

Figure 5. Transverse wave trajectories produced by cast TNT confined by one side (recording surface) only. Arrow indicates direction of detonation.

Figure 6. Transverse wave trajectories produced by nitromethane: acetone 70:30 (vol). Straight arrows indicate propagation direction. Dots correspond to transverse wave collision points. Insert is large-field view.

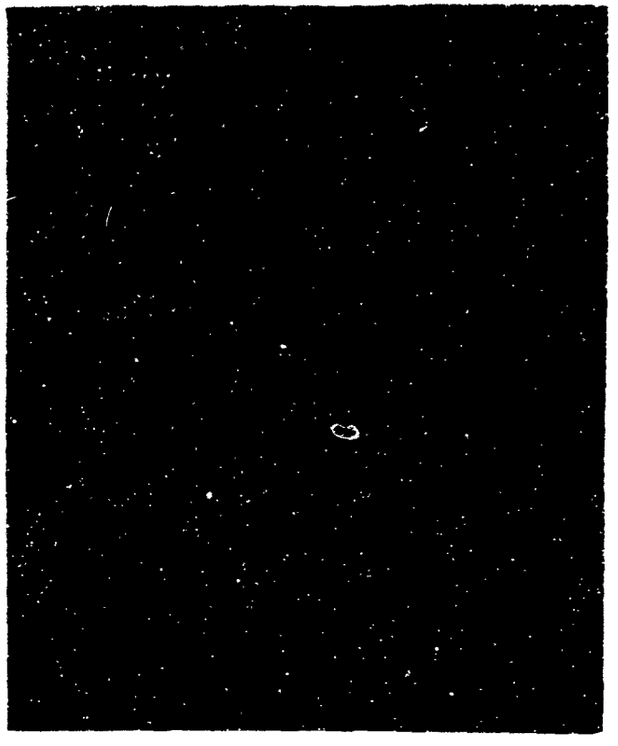

Figure 3. Wall writing pattern due to transverse waves in detonating nitromethane: acetone 80:20 (vol). Arrow indicates detonation propagation direction. Cell width ~ 0.6mm.

**Figure 4. Transverse wave trajectories produced by cast TNT confined on all sides. Arrow indicates direction of detonation. Cell width ~ 1.5mm.**

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